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The discovery of X-rays diffraction: from crystals to DNA. A case study to promote understanding of the nature of science and of its interdisciplinary character

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Abstract

The advantages of introducing history of science topics into the teaching of science has been advocated by a large number of scholars within the science education community. One of the main reasons given for using history of science in teaching is its power to promote understanding of the *nature of science* (NOS). In this respect, the historical case of X-rays diffraction, from the discovery of Max von Laue (1912) to the first X-rays diffraction photographs of DNA (1953), is a case in point for showing that a correct experimental strategy and a favourable theoretical context are not enough to make a scientific discovery.

Keywords

History of Science; Nature of Science; X-rays; X-rays diffraction; Wilhelm Conrad Röntgen; Max von Laue; DNA;

Introduction

The advantages of introducing history of science topics into the teaching of science has been advocated by a large number of scholars within the science education community (de Hosson & Schneeberger 2011, Leone 2014, Matthews 1994). As it was recently observed by Matthews (2012), one of the main reasons are the “cultural, educational, personal and scientific benefits of infusing the history and philosophy of science, into science programmes and curriculum; or in current terms, of teaching about the nature of science (NOS) while teaching science”. While there has been a long tradition advocating this approach, a number of open questions about NOS still exists. These questions deal with the optimal conditions for effective NOS teaching, the relationship between learning science and learning *about* science, and the issue of effectively measuring a NOS learning. Last but not least, there is an unsettled matter of definition arising from a lack of agreement in the science education community about what actually are the fundamentals of NOS (for a list of NOS elements according to some of the most influential authors in the field see Schwartz and Lederman 2008)

Notwithstanding these serious difficulties, and a conspicuous lack of experimental efforts to study the actual effectiveness of including history of science into science classes, curricula, and teacher education, a number of historical case studies have been studied with the goal of emphasizing its educational significance. These studies, rather than providing a shared list of

necessary and sufficient conditions for a practice to be scientific, identify “family resemblance of features that warrant different enterprises being called scientific” (Matthews 2012). In this respect, the discovery of X-rays diffraction by crystals, and some important outcomes like the emergence of X-rays spectroscopy and the discovery of DNA, are a case in point for showing that a correct experimental strategy and a favourable theoretical context are not enough to make a scientific discovery.

Max von Laue’s discovery of X-rays diffraction and the subsequent developments by William Henry Bragg and William Lawrence Bragg had been extensively discussed in Robotti (2012), to which we refer for a more detailed coverage of this topic.

The discovery of X-rays and of their nature

A major physics discovery occurred on November 8, 1895. For this discovery Wilhelm Conrad Röntgen, then professor of Physics at Wurzburg (Germany), was awarded the very first Nobel Prize in Physics (1901).

Röntgen had been studying the phenomenon of discharge of electricity through rarefied gases. By late 1860s it was known that if an evacuated glass tube is equipped with two electrodes and a voltage is applied, the glass opposite of the negative electrode (cathode) glows due to “cathode rays” (electrons) emitted from the cathode.

While working with a highly evacuated tube screened off by black paper, Röntgen discovered that a fluorescent screen brought near the tube, “lights up brilliantly and fluoresces, also if the screen is two meters away from the tube” (Röntgen 1895). This observation was entirely unexpected and soon became a classic case of accidental discovery.

Röntgen’s discovery was explained as the effect of a “new unknown form of invisible rays”. These new rays were shown to have a number of properties: they are emitted at the point of impact of cathode rays with wall of tube; travel in straight line; are highly penetrating; are able to impress a photographic plate; are neither reflected nor refracted. “For the sake of brevity” they were called “X-rays”. The new rays discovered by Röntgen were so spectacular that excited intense interest throughout the entire scientific world, and the first photographs obtained by them showed the by now reached ability to photograph the invisible.

From the year of their discovery to the first decade of twentieth century, X-rays were interpreted as electromagnetic waves of very short wavelength. However, in spite of this belief, no experimental demonstration of an analogy between light and X-rays existed. Furthermore, no reliable measurement of their wavelength was available. By 1912 both points were finally settled through the works, respectively, of Charles Grover Barkla and Arnold Sommerfeld. These accomplishments paved the way for the discovery of X-rays diffraction in crystals.

Röntgen had already attempted in 1895-97 to demonstrate the electromagnetic nature of X-rays by looking at an X-rays diffraction phenomenon by using both crystals and narrow slits. His attempts, however, got negative results (further efforts, to no avail as well, were made in 1899 by H. Haga and C.H. Wind, and in 1909 by B. Walter and R. Pohl, through wedge-shaped slits only a few microns wide). It was only in the 1906-1908 years that Barkla was able to provide strong evidence that X-rays consist of electromagnetic waves by studying the passage of X-rays through radiators. On the one hand, the scattered X-rays were indeed shown to be linearly polarized. On the other hand, heavy radiators were found to emit also a radiation “characteristic

of the radiator material” (the so-called “fluorescence radiation”), in analogy with Stokes law on light fluorescence (i.e. the radiation was emitted only when primary X-rays were harder than secondary ones) (Barkla 1906,1908).

As for the measurement of X-rays wavelength, it was taken in early 1912 by Sommerfeld, who had charged P.P. Koch to measure Walter and Pohl’s plates obtained with a new photometer just devised by Koch. The light curves, analyzed by Sommerfeld by means of his new theory on diffraction through wedge-shaped slits, showed a diffraction effect. Sommerfeld found indeed a considerable spectral range of the X-rays, whose center laid at a wavelength close to $4 \cdot 10^{-9}$ cm (Sommerfeld 1912).

The discovery of X-rays diffraction

In the fall of 1909 Max von Laue, former assistant to Max Planck in Berlin, went as *Privatdozent* to Munich at Sommerfeld’s Institute of Theoretical Physics. As he later wrote, “it turned out to be a matter of great good fortune that Sommerfeld passed to me the article ‘Wellenoptik’ (Wave optics) at that time to work upon for the *Encyclopedia of Mathematical Sciences*” (Laue 1915). In the effort of writing the entry he developed indeed a new theory of diffraction, valid not only for a linear grating (optical grating), but also for a cross-grating (lattice grids).

Laue’s attention in Munich was drawn constantly to the question of the nature of X-rays, “owing to the influence of Röntgen’s work at this University” (Röntgen had moved from Würzburg to Munich Institute of Experimental Physics in 1900) and as a consequence of “Sommerfeld’s active interest in X-rays” (Laue 1915). A further important circumstance was the presence in Munich of a third Institute, besides those headed by Röntgen and Sommerfeld: the Institute of Mineralogy and Crystallography. The idea of space-lattice arrangement of atoms was indeed widely known in Munich, mainly due to the role of P. Groth, director of this latest Institute.

In February 1912, P.P. Ewald, who was pursuing a doctorate on the optical properties of the lattice structure of crystals, under the guidance of Sommerfeld, asked Laue to help him to overcome some mathematical difficulties on the behavior of long electromagnetic waves in these structures (Ewald 1962). Having heard this, Laue “was suddenly struck by the obvious question” (Laue 1915), in view of his interests toward the X-rays: what behavior one might expect by *short* waves, like waves of X-rays wavelengths (10^{-9} cm), in a space lattice (constant of the order of 10^{-8} cm)?

Laue soon grasped that a crystal should behave for X-rays as a three-dimensional diffraction grating and that therefore space-lattice spectra would have to ensue.

At Laue suggestion, W. Friedrich (Sommerfeld’s assistant) and P. Knipping (a student graduating with Röntgen) volunteered to submit this possibility to experimental test.

By means of preliminary experiments with a copper sulphate crystal and a provisional apparatus, similar in principle to that used later, Friedrich and Knipping detected “a series of spots” together with a trace of the primary ray coming directly from the anticathode. The spots vanished if the same experiment was repeated with a “powdered” crystal, and similar results were obtained with other crystals. These results provided a strong support to Laue’s idea of X-rays diffraction by crystals.

Friedrich and Knipping later made use of an improved apparatus, where a widespread and fairly powerful tube was used (a Müller X-rays tube), and where the orientation of the crystal was sharply defined by an accurate goniometer (Figure 1).

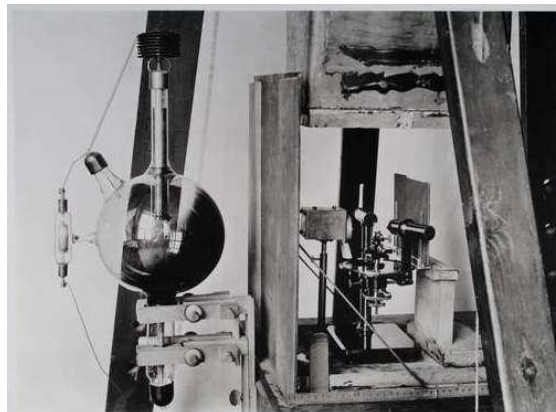


Figure 1. Friedrich and Knipping's apparatus (source: Deutsches Museum, Munich [Bragg Photos 4 nos. 86A-], Series 6. Accessed online via: William H. Bragg and William L. (Lawrence) Bragg: A guide to the research records of John Jenkin. <http://data.esrc.unimelb.edu.au/viewer/BRAG/item/BRAG00560/1>)

By this apparatus they obtained the first successful image of the X-rays diffraction in crystals, showing rings of fuzzy spots of elliptical shape, with the minor axis pointing to the overexposed centre of the black area produced by the primary ray (Figure 2) (Friedrich, Knipping and Laue 1912). To make the phenomenon more clear and easier to understand, they made the successful choice of using a cubic system crystal (the corresponding spatial lattice is the simplest possible), a zinc blende crystal, rather than the triclinic copper sulphate, previously used. Also, the sample was a plain parallel plate (10 x 10 x 0.5 mm) cut parallel to a face so that the X-rays struck the crystal perpendicularly to cube face.

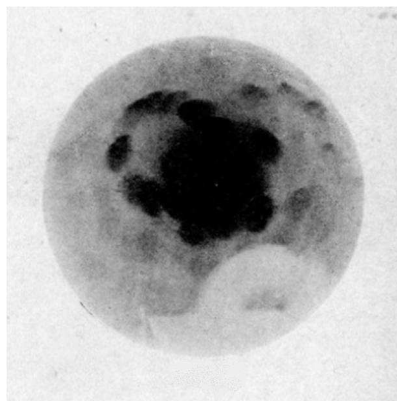


Figure 2. First successful photograph obtained by Friedrich and Knipping (source: Friedrich, Knipping, and Laue 1912)

Friedrich and Knipping found that the position of the spots was completely symmetrical in relation to the point of impact of the primary radiation. It was possible to see two pairs of planes of symmetry arranged perpendicular to each other. The fact that a completely fourfold symmetry is present on the plate was certainly the most beautiful demonstration of the space-lattice of the crystal, and of the fact that no other property than the space lattice is involved.

Other orientations of the zinc blende sample were used, e.g. if the zinc blende was irradiated along the threefold axis or the twofold axis, one could see the corresponding threefold or twofold symmetry. Also, additional samples were used, like rock salt and diamond plate.

To explain the images obtained, Laue developed the (yet nonexistent) diffraction theory for a space-lattice upon the basis of his article for the German Encyclopedia. He resumed his equation for a linear lattice and wrote it three times corresponding to the three periodicities of a space lattice. The observed rings of rays could thus be related to the cones of rays demanded separately by each of the three conditions of constructive interference. The spatial lattice considered was the most general one, that is the triclinic type (the edges of the elementary parallelepiped may thus have any lengths and be inclined at any angles to one other).

The comparison of the theory with the experimental data was done by Laue in the simplest case, namely that of zinc blende. He arrived at the conclusion that the diagrams were perfectly explainable on the assumption that the X-rays spectrum, rather than being continuous, contained only a number of discrete wavelengths and that these ones are responsible for the spots.

The discovery of X-rays diffraction in crystals benefited not only the lattice theory of crystals, but also the wave conception of X-rays. In fact, it was the triumph of this theory.

A new interpretation

William Henry Bragg, Cavendish Professor of Physics at the University of Leeds, tried to explain the effect observed by Friedrich, Knipping and Laue by his corpuscular hypothesis of X-rays. This approach, however, was soon abandoned and he, jointly with his son, the physicist William Lawrence Bragg from the University of Cambridge, adopted a wave conception of X-rays and came to the conclusion that Laue's was indeed a diffraction effect.

W.L. Bragg, however, suggested that Laue's explanation of the diffraction pattern was incorrect and unnecessarily complex. In order to explain the place of the spots, Laue was indeed forced to assume that only a few definite wavelengths are present in the incident beam. W.L. Bragg assumed instead that the X-rays beam is composed of a continuous range of wavelengths and that the diffraction patterns are due to an effect of reflection of the beam upon the crystal planes

After having observed that the points of a space lattice may be arranged in a series of planes, parallel and equidistant from each other (the simplest ones being the cleavage planes of the crystal), W.L. Bragg regarded "each interference maximum as due to the reflection of the X-rays in the systems of this plane" (Bragg 1913).

For a given wavelengths, the condition for the maxima was given by the law (eventually known as Bragg law)

$$n\lambda = 2d \sin \vartheta$$

where n is an integer, θ is the glancing angle, and d is the spacing of the planes.

Considered thus, W.L. Bragg wrote, “the crystal actually ‘manufactures’ light of definite wavelengths, much as ... a diffraction grating does” (Bragg 1913).

W.L. Bragg applied this new way of interpreting the diffraction pattern (that does not contradict Laue’s theory) to the zinc blende photographs analyzed by Laue. He assumed, following a suggestion by the chemist William Pope (Cambridge), that the zinc blende was a face centered cubic structure instead of, as assumed by Laue, a simple cubic structure (this assumption had indeed led Laue to estimate the cell size of the cubic lattice as smaller than $\sqrt[3]{4}$ and forced him to assume that only some wavelengths were present in the X-rays beam).

By this assumption, Bragg found that all the spots can be readily explained, also if other crystals were considered (figure 3).

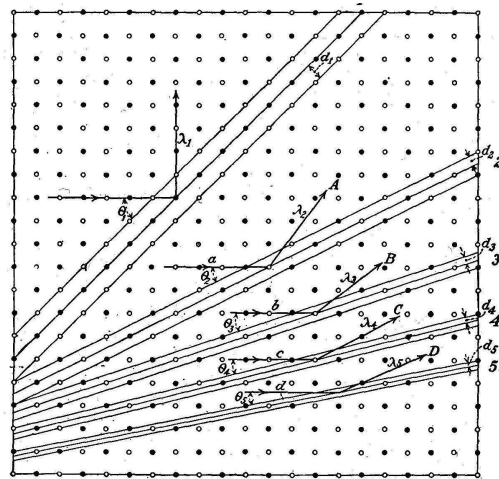


Figure 3. Schematic representation of the reflection by a crystal of rock salt (NaCl) of an heterochromatic X-rays incident beam (Richtmyer, F.K. (1934). *Introduction to modern physics*, 2nd edition, McGraw-Hill, New York)

On December 1912, W.L. Bragg carried out an experiment on a slip of mica and observed the specular reflection of the surface of the crystal.

This experiment opened up a period of close collaboration between father and son which is perhaps unique in the history of the science, both for its lasting intensity and the importance of the resulting discoveries.

In January 1913 W.H. Bragg succeeded in detecting the reflected rays with a ionization chamber, and two months later developed the first X-rays spectrometer, the instruments which for decades to come was the main tool for crystal structure analysis (it is an apparatus similar to an optical spectrometer in arrangement, an ionization chamber taking the place of a telescope) (Bragg and Bragg 1913).

By this new instrument, the Braggs measured the spectral distribution of the X-rays of their tube by using anticathodes of platinum, osmium, etc, and identified the K and L characteristic

radiations discovered by Barkla in 1911. These radiations, could be recognized also in the reflection from the faces of crystal.

Since April 1913 the focus of Bragg's work changed from the study of X-rays to the study of the structure of a crystal. By using the monochromatic K and L lines and measuring the angles at which these lines appeared after being reflected by the crystal, they could use the Bragg law on the reverse, that is to determine d and thus the structure of the crystal. By this method, several structures were confirmed and others discovered. For example, in July 1913 the Braggs studied the structure of diamond and discovered the tetrahedral arrangement of carbon atoms. At the end of the same year the crystal structure analysis became a standard procedure.

The importance, and also the history, of the discovery of X-rays diffraction is illustrated by the three Nobel prizes in Physics awarded between 1914 and 1917 for contributions within this field: to Laue in 1914 "for his discovery of the diffraction of X-rays by crystals", to the Braggs in 1915 "for their services in the analysis of crystal structure by means of X-rays", and to Barkla in 1917 "for his discovery of the characteristic Röntgen radiation of the elements".

Only a Nobel is oddly missing, notwithstanding a large number of nominations: Sommerfeld's one (Crawford 2002).

From X-rays spectroscopy to the DNA

In the period 1915-1920, following the fundamental Bragg's accomplishments, the X-rays spectroscopy laid the foundations for its successive development. Among the main results of this period are: accurate measurements of the X-rays wavelengths; analysis of large numbers of simple crystals by the new technique; discovery of a method to reliably measure the intensity of the reflected X-rays; measurement of Debye effect (that is the influence of temperature on the magnitude of X-rays reflection); development of Darwin's formulas for the intensity of X-rays reflection in crystals; understanding that each crystal diffraction corresponds to a Fourier component of the density of crystal; availability of a new set of crystal substances by the powder method, that in turn opens the way to the analysis of microcrystalline materials (Ewald 1962).

In the 1920s the X-rays spectroscopy becomes a quantitative science and is applied to increasingly complex structures, e.g. organic crystals. By studying naphthalene and anthracene W.L. Bragg showed in 1922 that the shapes of these molecules expected by organic chemistry fit well with actual measurements. In 1929 Kathleen Lonsdale discovered the structure of benzene and established that the derivatives of benzene are flat thereby putting an end to the mystery of aromatic hydrocarbons bonds. In 1925 the 2D Fourier analysis for crystal analysis is developed. In 1935 W.L. Patterson published a significant paper, introducing an important theoretical tool, the Patterson function, into the X-rays crystal structure analysis (Ewald 1962).

In late 1930s the first studies on biological macromolecules are pursued, and to 1930 are dated the first photographs of diffraction patterns from DNA fibres, obtained by Florence Bell, at the William Astbury Laboratory in Leeds. A new important photograph of DNA was taken in 1951, still at Astbury Laboratory, by Elwyn Beighton (Hall 2014).

However, the understanding of DNA structure required a theoretical discovery made in the same year by Linus Pauling and Robert Corey: the α -helix structure of proteins. In 1953 Pauling himself attempted to understand the DNA structure by this novel idea. His triple-helical model turned out, however, to be wrong (Pauling and Corey 1953).

The correct, *double helix*, model was suggested shortly later, still in 1953, by James Watson and Francis Crick with the help of the biophysicist Maurice Wilkins (Watson and Crick 1953; Wilkins, Stokes and Wilson 1953). This model was confirmed, and perhaps inspired, by an X-rays diffraction photograph of DNA obtained one year earlier (1952) by Raymond Gosling under Rosalind Franklin supervision at King's College of London (Franklin and Gosling 1953).

For this discovery Crick, Watson and Wilkins were awarded the Nobel Prize in Medicine 1962. Franklin untimely died in 1958 and was therefore ineligible for nomination to the Nobel Prize.

Her name, however, has lived on in history thanks to "Photo 51": a lasting symbol of the X-rays spectroscopy triumph (figure 4).



Figure 4. Gosling and Franklin's X-rays diffraction photograph of DNA (Photo 51) (source: https://askabiologist.asu.edu/Rosalind_Franklin-DNA).

Historical (and educational) conclusions

The above account shows that the discovery of X-rays diffraction was the final outcome of a lengthy process requiring a number of conditions: the success of the wave theory of X-rays mainly through Barkla's discovery of the fluorescence rays; the reliable estimate of X-rays wavelength; the emergence of an interest toward the crystal optics and the crystal lattice structure; and, finally, the development of an experimental expertise on X-rays and the commercial availability of fairly powerful X-rays tubes. All these conditions were met by 1912, particularly at the Sommerfeld's Department in Munich, where the scientific climate was favorable to Laue's discovery.

However, even if the search of X-rays diffraction was in the air in Munich, Laue was the one who had the idea that Nature gave us the right tool, that is a tool of resolving power high enough to diffract the X-rays, the crystal. Röntgen and others had looked for the diffraction by crystals, but to no avail. Laue succeeded where others had failed because he understood that the crystal may behave as a diffraction grating for X-rays. He knew what to look for and how to find it.

To make Laue's discovery a powerful experimental method, however, a new instrument was necessary, W.H. Bragg's X-rays spectrometer, and another fundamental idea was required, that is W.L. Bragg's idea that the diffraction might be seen as the internal reflection by the crystal planes.

These are all historiographical conclusions. However, these conclusions have also an educational significance. This case study shows indeed the presence of a number of the characteristics features of science (Matthews 2012).

The emergence in Munich of the discovery of X-rays diffraction emphasizes the social and cultural embeddedness of scientific knowledge. The way in which a crystal changes, in Laue's hands, into something new and unprecedented, that is a tool to observe the diffraction of X-rays, highlights other crucial features of science: the creative and imaginative nature of scientific knowledge, and the experimentation, or the Galilean importance of interfering with nature.

Lawrence Bragg's ways of looking at Laue's data shows the importance of idealization, or the fact that nature laws may not be always obvious in the immediate experience.

A final conclusion is in order. The discovery of X-rays diffraction, in turn, gave rise to the birth of a new field of science, the X-rays spectroscopy, that eventually led to one of the most significant discoveries of the 20th Century, the double helix model of the chromosome, thereby showing the role of models, and of their ubiquity in the history and current practice of science.

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